LA-UR-19-21423
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Title: Design principles for skyrmions in f-electron materials

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Intended for: LDRD ER

Issued: 2019-02-21

Objective

Spin textures with particle-like properties were recently discovered in bulk magnets [1]. Just like fundamental particles, these so-called skyrmions (see Fig. 1) can be created, stabilized and annihilated. Importantly, skyrmions can be easily moved through materials by current densities six orders of magnitude smaller than those of domain walls [2, 3]. Their particle-like properties arise from their unique topology and make them highly promising for applications in quantum information systems [4-8]. This will allow the U.S. to move beyond Moore's law—a grand challenge outlined both in LANL's materials strategy and a recent DOE Basic Research Needs report [9]. Despite this potential, progress towards efficient devices is currently limited because skyrmions are only observed in magnets lacking inversion symmetry [1], which drastically limits the number of skyrmion materials. *Here, based on our recent theoretical breakthrough in the modeling of mesoscale magnetic structures, we will pursue a new mechanism to stabilize skyrmions in f-electron materials for which LANL is a world leader. Our novel approach removes the constraint of broken inversion symmetry and also has advantages for applications. This approach can now be realized due to the unique co-location of multiscale modeling in T4 and synthesis and Hall measurement capabilities in MPA-CMMS.*

Capability: Innovation and Potential for Transformational Impact

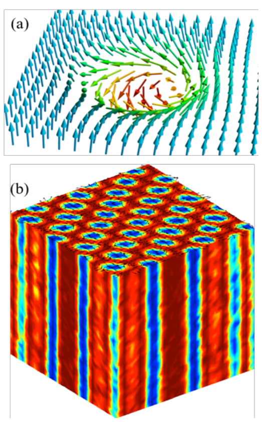

Figure 1, (a) spin profile at the cross-section of a skyrmion line. (b) Triangular skyrmion line lattice in *f*-electron system obtained by Monte Carlo simulations.

Currently, the only mechanism known to generate skyrmions in bulk magnets is the competition between a ferromagnetic exchange J, which favors parallel alignment of magnetic moments, and the Dzyaloshinskii-Moriya (DM) interaction of strength D, which favors canting of neighboring moments. These competing interactions result in a magnetic helix with mesoscopic period $\lambda=J/D$. The additional application of a small magnetic field stabilizes a superposition of three helices with propagation vectors $\boldsymbol{Q}_i$. This multi-$\boldsymbol{Q}$ state corresponds to a lattice of skyrmion lines with lattice parameter λ [see Fig. 1 (b)]. The DM interaction, however, is only allowed in materials that lack inversion symmetry, and the number of known materials is limited by this requirement. Available structural databases reveal that the number of materials without inversion symmetry is less than 20% of those with inversion symmetry. A powerful alternative is offered by *f*-electron materials for which it is well-establish that the competing RKKY interactions frequently stabilize helimagnetic order [10]. In a recent study, we show that, in the presence of multiple equivalent helical propagation vectors $\boldsymbol{Q}_i$ guaranteed by crystal symmetry, ***'f-electron skyrmions'*** can be stabilized via an easy-axis anisotropy [11,12]. Because the emergence of multi-$\boldsymbol{Q}$ order in *f*-electron materials is based on crystal symmetries, the magnetic lattice anisotropy that reflects the magnetoelastic coupling becomes important, in contrast to DM materials for which it is weak. It is well known that the RKKY interactions can stabilize magnetic helix with a large Q_i (small period), which therefore allows small skyrmions to be stabilized. Moreover, skyrmion lattices with different symmetries can be achieved by tuning the RKKY interaction profile [13], in contrast to DM materials for which only square and hexagonal symmetries are possible. *These properties result in unique advantages for skyrmions in f-electron materials:* (1) fewer symmetry constraints will greatly expand the number of available materials; (2) higher memory density will be enabled by their small size; and (3) the strong coupling of skyrmions to the underlying atomic lattice will enable straightforward manipulation of skyrmion properties by uniaxial strain. In addition, because easy-axis anisotropy can be controlled by uniaxial strain, one may stabilize *f*-electron skyrmions by strain engineering, or even control their trajectory in devices.

Mission Agility: Relevance and Leadership Potential

Understanding and anticipating new classes of functional materials is integral to national security in areas including energy security, sensing, and quantum computing. Success in identifying skyrmions in *f*-electron systems would have a large impact in the skyrmion and (more generally) spintronics research communities. It would also strengthen our leadership in the fields of both skyrmion and *f*-electron materials. By realizing design principles for skyrmions in *f*-electron materials, we will solve a key problem called out in the BASIC RESEARCH NEEDS WORKSHOP ON Quantum Materials for Energy Relevant Technology [9] and in the recent Request for Information called out by DARPA. Building upon the success in the proposed research, we will explore future funding opportunities within BES (early career) and LDRD in the areas of strongly correlated systems.

Research Approach

Theory: We will construct an effective magnetic Hamiltonian including RKKY interactions and magnetic anisotropy for candidate materials based on symmetry and available experimental data. We will use variational calculations and unbiased Monte Carlo simulations to obtain the magnetic phase diagram. Thermodynamic properties characteristic to skyrmion phase will be identified, which can be used to quickly screen possible skyrmion-hosting materials. Finally, we will calculate the novel properties of *f*-electron skyrmions relevant to applications. For example, we will study the influence of strain on the skyrmion phase, and the topological Hall effect under current drive. The numerical calculations will be performed using the supercomputers available through the Institutional Computing program.
Materials: Guided by our initial theoretical findings, we have identified several candidate materials, notably Nd [14, 15], $USb_{1-x}Te_x$ [16], $CeAuSb_2$ [17], CeAgBi2 [18], $DyRu_2Ge_2$ [19], NdIn3 [20], and CeFeB [21]. These materials show magnetic Bragg peaks consistent with multi-***Q*** ordering, but the associated magnetic structure remains unclear. We will grow high-quality single crystals of these materials using flux growth, vapor transport, Bridgman, and Czochralski.
Experiments: Initially, we will use bulk measurements to establish the phase diagram of candidate materials as function of temperature, magnetic field and strain to compare to theory. For example, ac susceptibility shows a characteristic two-peak structure in the imaginary part when a skyrmion phase is present. After this initial high-throughput screening, we will perform topological Hall measurement to verify the existence of topologically nontrivial skyrmion spin texture. Besides revealing the underlying magnetic textures, the Hall measurement gives additional insight into the interaction between conduction electrons and skyrmions, and the skyrmion dynamics. We will first focus on Nd single crystals, where the preliminary nuetron scattering measurement by our external collaborators suggests the existence of the skyrmion lattice (unpublished). We will measure the magnetization and electric Hall conductivity, which can be separated into several distinct contributions. We will look for the topological Hall conductivity, which could arise either from the topologically nontrivial electronic band structure or skyrmion lattice. These two contributions can be distinguished by combining magnetization and Hall measurements. Unixial strain cells will allow us to tune the easy-axis anisotropy *in-situ* and can be used to stabilize meta-stable skyrmions. Once we establish the existence of topological spin texture, we will collaborate with Marc Janoschek at Paul Scherrer Institute, who is an expert in neutron scattering study of mesoscopic spin texture, to map out the spin texture decisively.

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